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Magneto-Electric Jones Birefringence: A Bianisotropic Effect

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Abstract

In 1948, R. C. Jones showed that uniaxial media can in general show four different fundamental optical phenomena, each of which appears in refraction and absorption. Three of them are well-known: isotropic refraction and absorption, linear birefringence and dichroism, circular birefringence and dichroism. The fourth effect has remained unobserved so far. It represents an additional linear birefringence (and dichroism) with its fast and slow axes tilted by 45 degrees with respect to the axes of the standard linear birefringence. Jones birefringence should occur in several uniaxial crystal classes, and isotropic media subjected to external parallel magnetic and electric fields perpendicular to the direction of the light; $\Delta n_J = n_{+45^\circ} - n_{-45^\circ} = k_J \lambda \vec{E} \cdot \vec{B}$. We report the first experimental observation of the magneto-electric Jones birefringence, induced in liquids.

1. Introduction

Many linear optical effects in homogeneous uniaxial media are known, either intrinsic ones due to the symmetry properties of the medium, or effects induced by external influences like magnetic field, electric field, pressure etc. Jones developed a matrix formalism to classify these effects [1]. By a completeness argument, he deduced the existence of a fundamentally new effect in addition to the three known effects cited above. This fourth effect, appearing in refraction and absorption, is called Jones birefringence and Jones dichroism. Jones showed that this new effect can only exist in uniaxial media and that it represents an additional linear birefringence (with its corresponding dichroism), the optical axes of which bisect the optical axes of the standard linear birefringence. Later theoretical work on the basis of symmetry arguments [2] [3] and of generalized polarizability tensor calculations [2] showed that the Jones birefringence occurs intrinsically in many uniaxial crystal classes and can be induced in all media by simultaneously applying parallel magnetic and electric fields perpendicular to the direction of light propagation. This magneto-electric Jones birefringence Δn_J is predicted to be [2] [3]:

$$\Delta n_J \equiv n_{+45^\circ} - n_{-45^\circ} = k_J \lambda \, \vec{E} \cdot \vec{B} \tag{1}$$

where λ is the wavelength, $\vec{E}$ and $\vec{B}$ the external applied electric and magnetic fields. As the external fields can be modulated, the experimental search for the induced Jones birefringence allows high sensitivity (phase sensitive detection) and good discrimination against other effects, in particular the standard linear birefringence $\Delta n = n_{||} - n_{\perp}$ that always accompanies the Jones birefringence. In liquids and gases subjected to electric and magnetic fields, this standard birefringence is present in the form of the Cotton-Mouton effect ($\Delta n \propto B^2$) and the Kerr effect ($\Delta n \propto E^2$). These birefringences will in general be much stronger.

2. Theory

In the following modification of a much more complete treatment of Graham *et al.* [2] [6] we will show that Jones birefringence is a bianisotropic effect. In the expressions for $\vec{D}$ and $\vec{H}$ we retain the dominant terms of the light-matter interaction:

$$D_\alpha = \epsilon_0 E_\alpha + \alpha_{\alpha\beta} E_\beta + G_{\alpha\beta} B_\beta \qquad H_\alpha = \mu_0^{-1} B_\alpha - \chi_{\alpha\beta} B_\beta - \mathcal{G}_{\alpha\beta} E_\beta \tag{2}$$

In a liquid subjected to parallel electric and magnetic fields along the x-axis, E_{ext}, B_{ext}, the tensors become field distorted, for example: $G_{\alpha\beta}(E_{ext}, B_{ext}) = G_{\alpha\beta} + \frac{\partial G_{\alpha\beta}}{\partial B_{ext}} B_{ext} + \frac{\partial G_{\alpha\beta}}{\partial E_{ext}} E_{ext} + \cdots$. The multipole polarization densities are statistical averages over the molecular orientations in the external fields. We consider only polar diamagnetic molecules and only averages proportional to $E_{ext} \cdot B_{ext}$ [2]. The dominant polar contribution is given by:

$$\langle G_{\alpha\beta}(E_{ext}, B_{ext}) \rangle = -\frac{E_{ext} B_{ext}}{kT} \langle \mu_x \frac{\partial \mathcal{G}_{\alpha\beta}}{\partial B_{ext}} \rangle \tag{3}$$

Analogous for $\langle \mathcal{G}_{\alpha\beta} \rangle$, but $\langle \epsilon_0 \delta_{\alpha\beta} + \alpha_{\alpha\beta} \rangle = \epsilon_0 \delta_{\alpha\beta}$ and $\langle \mu_0^{-1} \delta_{\alpha\beta} + \chi_{\alpha\beta} \rangle = \mu_0^{-1} \delta_{\alpha\beta}$. We rewrite:

$$D_\alpha = \epsilon_0 \delta_{\alpha\beta} E_\beta + \frac{N E_{ext} B_{ext}}{kT} \langle \mu_x \frac{\partial \mathcal{G}_{\alpha\beta}}{\partial B_{ext}} \rangle B_\beta \qquad H_\alpha = \mu_0^{-1} \delta_{\alpha\beta} B_\beta - \frac{N E_{ext} B_{ext}}{kT} \langle \mu_x \frac{\partial \mathcal{G}_{\alpha\beta}}{\partial B_{ext}} \rangle E_\beta \tag{4}$$

We assume plane wave eigenmodes $E_\alpha = E_{o_\alpha} e^{-i\omega(t - \frac{n}{c} z)}$ and use the Maxwell equations $E_\alpha = \frac{n}{c} \epsilon_{\alpha z \beta} B_\beta$ and $D_\alpha = -\frac{n}{c} \epsilon_{\alpha z \beta} H_\beta$. This leads, with $\frac{\partial \mathcal{G}_{\alpha\beta}}{\partial B_{ext}} = \frac{\partial G_{\beta\alpha}}{\partial B_{ext}}$ [2], to the wave equation:

$$\begin{pmatrix} n^2 & -\frac{n}{c\epsilon_0} \frac{N E_{ext} B_{ext}}{kT} \langle \mu_x (\frac{\partial G_{yy}}{\partial B_{ext}} - \frac{\partial G_{xx}}{\partial B_{ext}}) \rangle \\ -\frac{n}{c\epsilon_0} \frac{N E_{ext} B_{ext}}{kT} \langle \mu_x (\frac{\partial G_{yy}}{\partial B_{ext}} - \frac{\partial G_{xx}}{\partial B_{ext}}) \rangle & n^2 \end{pmatrix} \begin{pmatrix} E_{o_x} \\ E_{o_y} \end{pmatrix} = \begin{pmatrix} E_{o_x} \\ E_{o_y} \end{pmatrix} \tag{5}$$

The eigenvectors are linearly polarized light at $\pm$ 45° with $\Delta n_J = \frac{1}{c\epsilon_0} \frac{N E_{ext} B_{ext}}{kT} \langle \mu_x (\frac{\partial G_{yy}}{\partial B_{ext}} - \frac{\partial G_{xx}}{\partial B_{ext}}) \rangle$.

3. Experiment

The experimental setup is shown in figure 1. It is a modification of a setup to measure magnetic linear birefringence, the Cotton-Mouton effect [4]. It consists of a HeNe-laser ^{L}, polarizer ^{P}, photoelastic modulator ^{PEM}, Pockels' cell ^{PC}, Fresnel rhomb ^{FR}, sample cell ^{S}, analyzer ^{A} and photodiode ^{PD}. Phase-sensitive feedback loop drives the Pockels' cell to compensate the sample birefringence. Sample lengths varied from 5 to 25 mm. Applied are a static magnetic field $\vec{B}$ and a low frequency alternating electric field $\vec{E} \cdot \cos \Omega t$. A phase sensitive detection of the resulting birefringence at the electric field frequency Ω ($\approx 60 s^{-1}$) is performed. The angle ϕ of the polarization of the light incident on the sample with respect to the magnetic field can be chosen with the Fresnel rhomb. The angle θ between $\vec{B}$ and $\vec{E}$ can be chosen by rotating the electrode assembly. By a proper choice of ϕ and θ and the external fields, the setup can measure electric linear (Kerr), magnetic linear (Cotton-Mouton) and magneto-electric Jones birefringence. The resolution is $\Delta n_J \approx 2 \cdot 10^{-12}$ with applied fields of 17 T and 2,5 $\cdot 10^5 \frac{\text{V}}{\text{m}}$ and a pathlength of 25 mm.

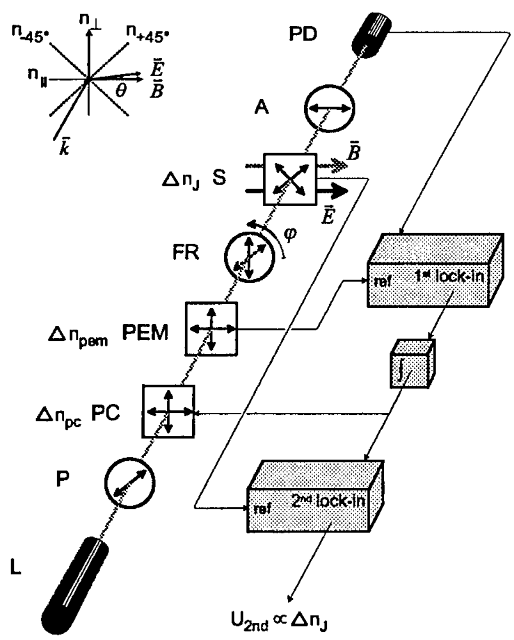

Figure 1: Experimental setup

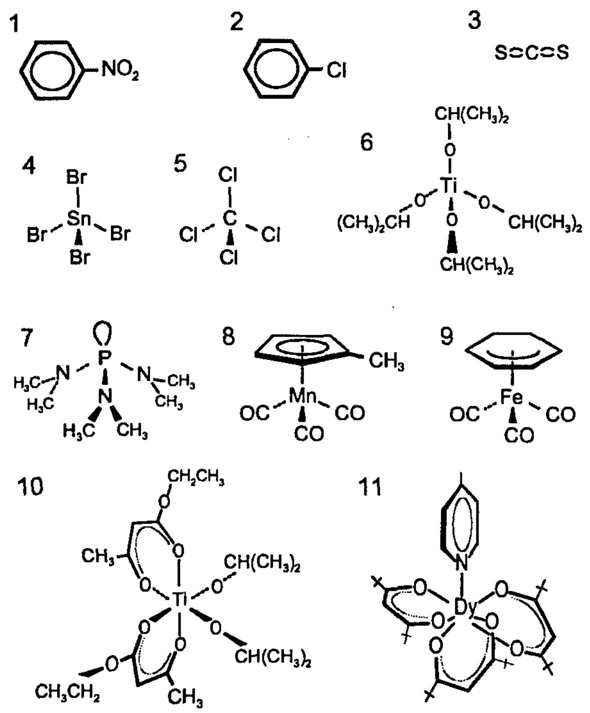

Figure 2: Investigated molecules

The samples were pure molecular liquids or concentrated molecular solutions and non-absorbing at the laser wavelength of 632 nm. They were selected based on the presence of low-lying, high oscillator strength optical transitions and the possibility to have high concentrations. Dipolar **1**,**2**, quadrupolar **3**, tetrahedral **4**-**9** and octahedral **10**,**11** molecules (the symmetries being only approximate) have been examined, see figure 2. Only **8**, **9** and **10** showed a significant Jones birefringence. Typical results for **8** are shown in figure 3, demonstrating explicitly the linear dependence of Δn_J on E, B and $\cos\theta$, thereby proving the existence of the Jones birefringence and confirming equation 1. The table summarizes the results for the molecules **8**, **9** and **10** (the electric field and Δn_J in the last column are rms values). It was further checked that the observed Δn_J is independent of the sample length. The temperature dependence of Δn_J for molecule **8** was measured to be $k_J(T) \propto T^{-x}, x = 0.8 \pm 0.2$. This is consistent with the alignment of a permanent molecular dipole moment by an external field, which would give $x = 1$. We suppose this moment to be the static electric dipole moment.

A further test for the validity of our experimental results consisted of measuring the magneto-electric linear birefringence of **8** and **10** in crossed magnetic and electric fields, both perpendicular to the direction of light propagation. This birefringence, which has the same optical axes as the Kerr and Cotton-Mouton effects, was predicted to have the same magnitude as the Jones birefringence [3] [5]. We found this to be indeed the case.

	μ_e [D]	μ_m [μ_B]	k_J $\left[\frac{1}{\mathrm{VT}}\right]$	$\Delta n_J^{17\,\mathrm{T}/1{,}9\cdot 10^5\,\frac{\mathrm{V}}{\mathrm{m}}}$
8	≈ 4	1,7	$4,7 \cdot 10^{-11}$	$9,6 \cdot 10^{-11}$
9	≈ 4	≈ 3	$2,2 \cdot 10^{-11}$	$4,5 \cdot 10^{-11}$
10	$\neq 0$	≈ 0	$5,1 \cdot 10^{-12}$	$1,0 \cdot 10^{-11}$

We can heuristically summarize our results by noting that a relatively large Jones birefringence is observed in molecules having a low-lying strong charge transfer transition of approximately octupolar symmetry and a permanent electric dipole moment. Note that all liquids must exhibit the magneto-electric Jones birefringence, but the effect is below our detection limit for the other molecules.

Graham and Raab estimated the strength of the Jones birefringence for spherical atoms. They found the following relation between Jones, Cotton-Mouton and Kerr birefringence [2]:

$$\eta \equiv \left| \frac{\Delta n_J}{\sqrt{\Delta n_K \Delta n_{CM}}} \right| = \left| \frac{k_J}{\sqrt{k_K\ k_{CM}}} \right| = 0.016 \qquad (6)$$

The results of Ross *et al.* [3] would imply that η is of the order of α, the fine structure constant (≈ 0.0073). The values we have observed so far are $\eta = 0.0036$ for **8** and **10** and $\eta = 0.0019$ for **9** . This is within one order of magnitude of both predictions. However, values for η that are at least two orders of magnitude smaller than this were found for other molecules. The relation between η and the molecular structure is not yet understood.

Estimates of the absolute strength of Jones birefringence have been made for hydrogen atoms [5]. $k_J = 6 \cdot 10^{-17} \frac{1}{\mathrm{VT}}$ was calculated at 1 atm. pressure, which translates to $k_J \approx 10^{-14} \frac{1}{\mathrm{VT}}$ for the densities of our molecular liquids. Electrostatic alignment of permanent dipole moments at room temperature increases this by two orders of magnitude [2], thus $k_J \approx 10^{-12} \frac{1}{\mathrm{VT}}$. Resonance enhancement due to the low-lying optical transitions may give another order of magnitude, leading to an estimate of $k_J \approx 10^{-11} \frac{1}{\mathrm{VT}}$, which is in reasonable agreement with our experimental results for k_J on **8**, **9** and **10**. Empirically, this extrapolation seems only to be valid for those molecules that have optical transitions that involve truly three-dimensional motion of electrons, as is also the case for the hydrogen atom.

4. Conclusion

We have for the first time experimentally observed the Jones birefringence, induced by an electric and a magnetic field in molecular liquids. This observation provides the final validation of the Jones formalism in polarization optics. Our results confirm all qualitative predictions made for the effect. However our understanding of the relation between Jones birefringence and molecular structure is still incomplete.

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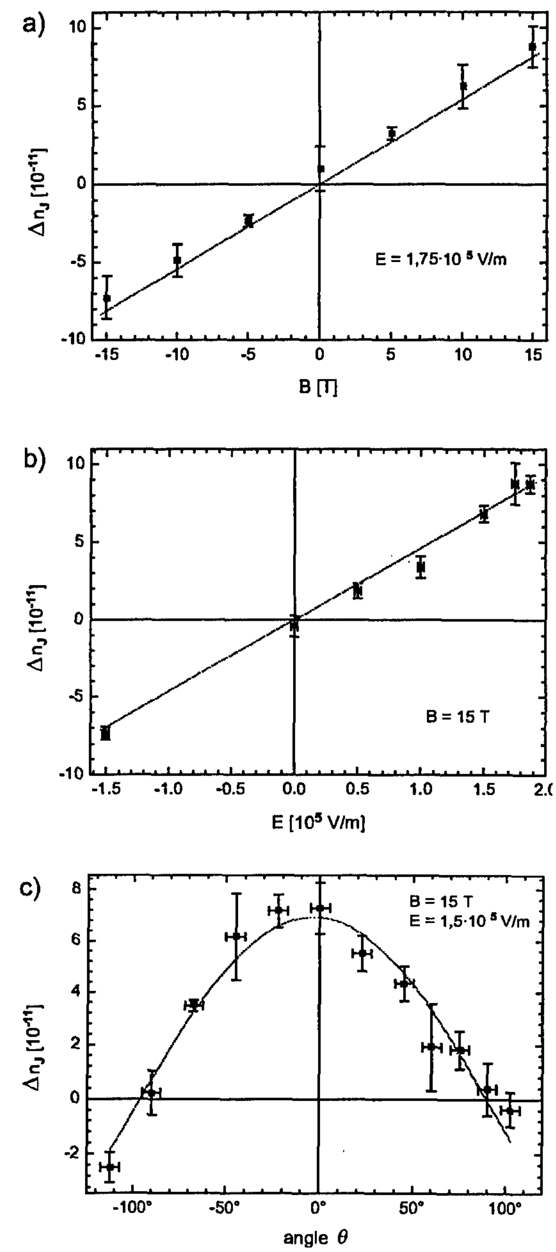

Figure 3: Characteristic dependencies

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